# Relaxation of the Electron Spin in Quantum Dots Via One- and Two-Phonon Processes

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We have studied direct and Raman processes of the decay of electron spin states in a quantum dot via radiation of phonons corresponding to elastic twists. Universal dependence of the spin relaxation rate on the strength and direction of the magnetic field has been obtained in terms of the electron gyromagnetic tensor and macroscopic elastic constants of the solid.

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## I. INTRODUCTION

Electron spin relaxation in solids is related to such important applied problems as electron spin resonance and quantum computing. Many interactions contribute to the relaxation time of the electron spin in a semiconductor quantum dot. In principle, all of them can be eliminated (such as, e.g., impurities, nuclear spins, etc.) but the interaction with phonons cannot. Thus, spin-phonon interaction provides the most fundamental upper bound on the lifetime of electron spin states. The existing methods of computing electron spin-phonon rates in semiconductors rely upon phenomenological models of spin-orbit interaction, see, e.g., Refs. [1, 2, 3, 4, 5, 6, 7, 8, 9, 10]. These models contain unknown constants that must be obtained from experiment. Meantime, the spin-orbit coupling determines the difference of the electron g-tensor from the unit tensor. The question then arises whether the effect of the spin-orbit coupling on spin-phonon relaxation can be expressed via the difference between the electron gyromagnetic tensor $g_{\alpha\beta}$ and the vacuum tensor $g_0\delta_{\alpha\beta}$. Since $g_{\alpha\beta}$ can be measured independently, this would enable one to compare the computed relaxation rates with experiment without any fitting parameters. In this paper we show that this can be done under certain reasonable simplifying assumptions. We obtain spin-phonon relaxation rates due to direct one-phonon processes and due to two-phonon Raman processes.

Zeeman interaction of the electron with an external magnetic field, $\mathbf{H}$, is given by the Hamiltonian

$$\hat{H}_Z = -\mu_B\, g_{\alpha\beta}\, S_\alpha H_\beta\ , \tag{1}$$

where $\mu_B$ is the Bohr magneton and $\mathbf{S} = \boldsymbol{\sigma}/2$ is the dimensionless electron spin, with $\sigma_\alpha$ being Pauli matrices. One can choose the axes of the coordinate system along the principal axes of the tensor $g_{\alpha\beta}$. Then, $g_{\alpha\beta} = g_\alpha\delta_{\alpha\beta}$. Perturbation of Eq. (1) by phonons has been studied in the past [3, 4, 6] by writing all terms of the expansion of $g_{\alpha\beta}$ on the strain tensor, $u_{\alpha\beta}$, permitted by symmetry. This gives spin-phonon interaction of the form $A_{\alpha\beta\gamma\rho}u_{\alpha\beta}S_\gamma B_\rho$ with unknown coefficients $A_{\alpha\beta\gamma\rho}$. To avoid this uncertainty we limit our consideration to local rotations generated by transverse phonons. The argument for doing this is three-fold. Firstly, the rate of the transition accompanied by the emission or absorption of a phonon is inversely proportional to the fifth power of the sound velocity [11]. Since the velocity of the transverse sound is always smaller than the velocity of the longitudinal sound [12], the transverse phonons must dominate the transitions. Secondly, we notice that interaction of the electron spin with a local elastic twist generated by a transverse phonon does not contain any unknown constants. Consequently, it gives parameter-free lower bound on the spin relaxation. Thirdly, for a dot that is sufficiently rigid to permit only tiny local rotations as a whole under an arbitrary elastic deformation, the emission or absorbtion of a quantum of the elastic twist is the only spin-phonon relaxation channel.

The angle of the local rotation of the crystal lattice in the presence of the deformation, $\mathbf{u}(\mathbf{r})$, is given by [12]

$$\delta\boldsymbol{\phi} = \frac{1}{2}\boldsymbol{\nabla}\times\mathbf{u}\ . \tag{2}$$

As stated above, the gyromagnetic tensor $g_{\alpha\beta}$ is determined by the local environment of the quantum dot. In the presence of long wave deformations of the lattice, the whole environment is rotated so that the gyromagnetic tensor becomes

$$g_{\alpha\beta} = \mathbb{R}_{\alpha\alpha'}\mathbb{R}_{\beta\beta'}g_{\alpha'\beta'}\ , \tag{3}$$

where $\mathbb{R}_{\alpha\beta}$ is the $3\times 3$ rotation matrix given by $\delta\boldsymbol{\phi}(\mathbf{r})$. One can thus write the total Hamiltonian in the form

$$\hat{\mathcal{H}} = \hat{\mathcal{H}}'_Z + \hat{\mathcal{H}}_{ph}\ , \tag{4}$$

where

$$\hat{\mathcal{H}}'_Z = -\mu_B g_{\alpha'\beta'}\left(\mathbb{R}^{-1}_{\alpha'\alpha}S_\alpha\right)\left(\mathbb{R}^{-1}_{\beta'\beta}H_\beta\right)\ . \tag{5}$$

and $\hat{\mathcal{H}}_{ph}$ describes free harmonic phonons. In the above formulae, $\delta\boldsymbol{\phi}$ must be understood as an operator. Indeed, from the canonical quantization of phonons and Eq. (2) one obtains

$$\delta\boldsymbol{\phi} = \sqrt{\frac{\hbar}{\mathbf{8\rho V}}}\sum_{\mathbf{k\lambda}}\frac{[\mathbf{ik}\times\mathbf{e_{k\lambda}}]\,\mathbf{e}^{\mathbf{ik\cdot r}}}{\sqrt{\boldsymbol{\omega}_{\mathbf{k\lambda}}}}\left(\mathbf{a_{k\lambda}}+\mathbf{a}^\dagger_{\mathbf{-k\lambda}}\right) \tag{6}$$

where $\rho$ is the mass density, $V$ is the volume of the crystal, $\mathbf{e}_{\mathbf{k}\lambda}$ are unit polarization vectors, $\lambda = t_1, t_2, l$ denotes polarization, and $\omega_{k\lambda} = v_\lambda k$ is the phonon frequency.

## II. DIRECT PROCESSES

In order to account for spin transitions accompanied by the emission or absorption of one phonon one needs to consider terms up to first order in phonon amplitudes. Therefore, with the help of the expansion of the rotation matrix to the first order in $\delta\boldsymbol{\phi}$, $\mathbb{R}_{\alpha\beta} = \delta_{\alpha\beta} - \epsilon_{\alpha\beta\gamma}\delta\phi_\gamma$, we obtain the full Hamiltonian

$$\hat{\mathcal{H}} = \hat{\mathcal{H}}_0 + \hat{\mathcal{H}}_{s-ph}\,, \qquad \hat{\mathcal{H}}_0 = \hat{\mathcal{H}}_Z + \hat{\mathcal{H}}_{ph}\,, \tag{7}$$

where

$$\hat{\mathcal{H}}_{s-ph} = -\mu_B\epsilon_{\alpha\beta\gamma}(g_\alpha - g_\beta)H_\beta\delta\phi_\gamma S_\alpha\,. \tag{8}$$

Spin-phonon transitions occur between the eigenstates of $\hat{\mathcal{H}}_0$, which are direct products of spin and phonon states

$$|\Psi_\pm\rangle = \left|\psi_\pm\right\rangle \otimes \left|\phi_\pm\right\rangle\,. \tag{9}$$

Here $\left|\psi_\pm\right\rangle$ are the eigenstates of $\hat{\mathcal{H}}_Z$ with energies $E_\pm$ and $\left|\phi_\pm\right\rangle$ are the eigenstates of $\hat{\mathcal{H}}_{\rm ph}$ with energies $E_{\rm ph\pm}$. Spin-phonon processes conserve energy, $E_+ + E_{\rm ph+} = E_- + E_{\rm ph-}$. For $\hat{\mathcal{H}}_{s-ph}$ of Eq. (7), the states $\left|\phi_\pm\right\rangle$ differ by one emitted or absorbed phonon with wave vector $\mathbf{k}$. We will use the following designations

$$\left|\phi_+\right\rangle \equiv |n_{\mathbf{k}}\rangle\,,\ \left|\phi_-\right\rangle \equiv |n_{\mathbf{k}}+1\rangle\,. \tag{10}$$

To obtain the relaxation rate one can use the Fermi golden rule. The matrix element corresponding to the decay of the spin $|\Psi_+\rangle \to |\Psi_-\rangle$ can be evaluated with the help of Eqs. (8) and (6) we obtain

$$\langle\Psi_-|\hat{\mathcal{H}}_{s-ph}|\Psi_+\rangle = \frac{\hbar}{\sqrt{V}}\sum_{\mathbf{k}\lambda} V_{\mathbf{k}\lambda}\langle n_{\mathbf{k}'}+1|a_{\mathbf{k}\lambda} + a^\dagger_{-\mathbf{k}\lambda}|n_{\mathbf{k}'}\rangle$$
$$V_{\mathbf{k}\lambda} \equiv \frac{e^{i\mathbf{k}\cdot\mathbf{r}}}{\sqrt{8\rho\hbar\omega_{\mathbf{k}\lambda}}}\,\mathbf{K}\cdot[\mathbf{k}\times\mathbf{e}_{\mathbf{k}\lambda}] \tag{11}$$

where the components of vector $\mathbf{K}$ are given by

$$K_\gamma \equiv -\mu_B\epsilon_{\alpha\beta\gamma}(g_\alpha - g_\beta)H_\beta\langle\psi_-|S_\alpha|\psi_+\rangle\,. \tag{12}$$

Note that only the transverse phonons are considered in the summations. For the direct process, the decay rate, $\Gamma_D$, is, then,

$$\Gamma_D = \Gamma_0\coth\left(\frac{\hbar\omega_0}{2k_BT}\right)\,. \tag{13}$$

where

$$\hbar\omega_0 \equiv E_+ - E_- = \mu_B\left(\sum_\gamma g_\gamma^2 H_\gamma^2\right)^{1/2} \tag{14}$$

is the distance between the two spin levels, and

$$\Gamma_0 = \frac{1}{V}\sum_{\mathbf{k}\lambda}|V_{\mathbf{k}\lambda}|^2 2\pi\delta(\omega_{\mathbf{k}\lambda} - \omega_0)\,. \tag{15}$$

Using Eq. (11) and replacing $\sum_{\mathbf{k}}$ by $(V/(2\pi)^3)\int d^3k$, one obtains

$$\Gamma_0 = \frac{1}{12\pi\hbar}\frac{|\mathbf{K}|^2\omega_0^3}{\rho v_t^5}\,, \tag{16}$$

where $v_t$ is the velocity of the transverse sound. A straightforward calculation of the spin matrix elements yields

$$|\mathbf{K}|^2 = \frac{\mu_B^2}{8}\sum_{\alpha\beta=x,y,z}(g_\alpha - g_\beta)^2 \times\left[B_\alpha^2 + B_\beta^2 - \frac{(g_\alpha+g_\beta)^2H_\alpha^2H_\beta^2}{\sum_\gamma(g_\gamma H_\gamma)^2}\right]. \tag{17}$$

Then, the decay rate can be written in the final form

$$\Gamma_D = \frac{\hbar}{3\pi\rho}\left(\frac{\mu_B H}{\hbar v_t}\right)^5 F_T(\mathbf{n})\,, \tag{18}$$

where $\mathbf{n} \equiv \mathbf{H}/H$ and

$$F_T(\mathbf{n}) = \left(\sum_\gamma g_\gamma^2 n_\gamma^2\right)^{3/2}\coth\left[\frac{\mu_B H}{2k_BT}\left(\sum_\gamma g_\gamma^2 n_\gamma^2\right)^{1/2}\right] \times\frac{1}{32}\sum_{\alpha\beta}(g_\alpha - g_\beta)^2\left[n_\alpha^2 + n_\beta^2 - \frac{(g_\alpha+g_\beta)^2 n_\alpha^2 n_\beta^2}{\sum_\gamma(g_\gamma n_\gamma)^2}\right]. \tag{19}$$

If the field is directed along the $Z$-axis, Eq. (19) simplifies to

$$F_T(\mathbf{e}_z) = \frac{g_z^3}{32}[(g_z - g_x)^2 + (g_z - g_y)^2]\coth\left(\frac{g_z\mu_B B}{2k_BT}\right). \tag{20}$$

When components of $g_{\alpha\beta}$ are of order unity and $k_BT \gtrsim \mu_B H$, then $\Gamma_D \sim (k_BT/\hbar)(\mu_B H/E_t)^4$, where we have introduced $E_t \equiv (\hbar^3\rho v_t^5)^{1/4} \sim 10^2$K.

## III. RAMAN PROCESSES

Spin-lattice relaxation by Raman scattering is a two phonon mechanism consisting of a spin transition accompanied by the absorption of a phonon and the emission of another phonon of different frequency. In spite of being a second order process, its contribution can be very important, since the phase space of the phonons triggering the transition is not limited to the distance between the levels as in the direct case. To describe such processes, we need to consider terms up to second order in phonon amplitudes in the Hamiltonian. To this end we expand the $3\times 3$ rotation matrix to second order in $\delta\boldsymbol{\phi}$

$$\mathbb{R}_{\alpha\beta} = \delta_{\alpha\beta} - \epsilon_{\alpha\beta\gamma}\delta\phi_\gamma + \frac{1}{2}\left[\delta\phi_\alpha\delta\phi_\beta - \delta_{\alpha\beta}(\delta\boldsymbol{\phi})^2\right] \tag{21}$$

and obtain the full Hamiltonian from Eqs. (4) and (5)

$$\hat{\mathcal{H}} = \hat{\mathcal{H}}_0 + \hat{\mathcal{H}}_{s-ph}\,, \qquad \hat{\mathcal{H}}_{s-ph} = \hat{\mathcal{H}}^{(1)}_{s-ph} + \hat{\mathcal{H}}^{(2)}_{s-ph}\,, \tag{22}$$

where $\hat{\mathcal{H}}^{(1)}_{s-ph}$ is given by Eq. (8) and

$$\begin{aligned}\hat{\mathcal{H}}^{(2)}_{s-ph} &= -\frac{1}{2}\mu_B(g_\alpha+g_\beta)S_\alpha H_\beta \delta\phi_\alpha\delta\phi_\beta \\ &- \mu_B g_\alpha(\mathbf{S}\times\delta\boldsymbol{\phi})_\alpha(\mathbf{H}\times\delta\boldsymbol{\phi})_\alpha\,. \end{aligned} \tag{23}$$

Again, we will study the spin-phonon transitions between the eigenstates of $\hat{\mathcal{H}}_0$, $|\Psi_\pm\rangle = |\psi_\pm\rangle\otimes|\phi_\pm\rangle$. Here, again,$|\psi_\pm\rangle$ are the eigenstates of $\hat{\mathcal{H}}_Z$ with energies $E_\pm$ and $|\phi_\pm\rangle$ the eigenstates of $\hat{\mathcal{H}}_{ph}$ with energies $E_{ph\pm}$. We consider Raman processes, in which a phonon with wave vector $\mathbf{k}$ is absorbed and a phonon with a wave vector $\mathbf{q}$ is emitted. We will use the following designations for the phonon states

$$|\phi_+\rangle \equiv |n_\mathbf{k}, n_\mathbf{q}\rangle, \qquad |\phi_-\rangle \equiv |n_\mathbf{k}-1, n_\mathbf{q}+1\rangle\,. \tag{24}$$

To obtain the relaxation rate of the transition $|\Psi_+\rangle \to |\Psi_-\rangle$ one needs to evaluate the matrix element of the process, which is the sum of the matrix element with $\hat{\mathcal{H}}^{(2)}_{s-ph}$ and that with $\hat{\mathcal{H}}^{(1)}_{s-ph}$ in the second order: $M_R = M^{(2)}_R + M^{(1+1)}_R$, where

$$M^{(2)}_R = \langle\Psi_-|\,\hat{\mathcal{H}}^{(2)}_{s-ph}\,|\Psi_+\rangle \tag{25}$$

and

$$\begin{aligned}M^{(1+1)}_R = &\sum_{\xi=\pm}\frac{\left\langle\Psi_-\left|\hat{\mathcal{H}}^{(1)}_{\rm s-ph}\right|\Psi_\xi\right\rangle\left\langle\Psi_\xi\left|\hat{\mathcal{H}}^{(1)}_{\rm s-ph}\right|\Psi_+\right\rangle}{E_+ + \hbar\omega_\mathbf{k} - E_\xi} \\ &+\sum_{\xi=\pm}\frac{\left\langle\Psi_-\left|\hat{\mathcal{H}}^{(1)}_{\rm s-ph}\right|\Psi_\xi\right\rangle\left\langle\Psi_\xi\left|\hat{\mathcal{H}}^{(1)}_{\rm s-ph}\right|\Psi_+\right\rangle}{E_+ - E_\xi - \hbar\omega_\mathbf{q}}. \end{aligned} \tag{26}$$

The intermediate phonon states are $|n_\mathbf{k}-1, n_\mathbf{q}\rangle$ in the first term and $|n_\mathbf{k}, n_\mathbf{q}+1\rangle$ in the second term.

Raman processes may dominate over direct processes for a small energy difference between the spin states, $\hbar\omega_0 \ll k_BT$. Thus we will consider terms to the lowest order in H. For simplicity we will study the case where the field is directed along the $Z$-axis. Then the matrix element becomes,

$$\begin{aligned}M_R &= M^{(2)}_R = \frac{\mu_B H}{4}\left[(2g_y - g_x - g_z)\tilde{M}^{xz}_{ph-R}\right. \\ &- \left. i(2g_x - g_y - g_z)\tilde{M}^{yz}_{ph-R}\right], \end{aligned} \tag{27}$$

where $\tilde{M}^{\alpha\beta}_{ph-R} = M^{\alpha\beta}_{ph-R} + M^{\beta\alpha}_{ph-R}$ with

$$\begin{aligned}M^{\alpha\beta}_{ph-R} &= \langle n_\mathbf{q}+1\,|\delta\phi_\alpha|\,n_\mathbf{q}\rangle\,\langle n_\mathbf{k}-1\,|\delta\phi_\beta|\,n_\mathbf{k}\rangle \\ &= \frac{\hbar^2}{8\rho V}\frac{[\mathbf{k}\times\mathbf{e}_{\mathbf{k}\lambda_\mathbf{k}}]_\alpha\,[\mathbf{q}\times\mathbf{e}_{\mathbf{q}\lambda_\mathbf{q}}]_\beta}{\sqrt{\hbar\omega_{\mathbf{k}\lambda_\mathbf{k}}\hbar\omega_{\mathbf{q}\lambda_\mathbf{q}}}}\sqrt{(n_\mathbf{q}+1)\,n_\mathbf{k}} \end{aligned} \tag{28}$$

The Raman rate of the transition $|\Psi_+\rangle \to |\Psi_-\rangle$ can be obtained by using the Fermi golden rule,

$$\begin{aligned}\Gamma_R &= \left[(2g_y - g_x - g_z)^2 + (2g_x - g_y - g_z)^2\right] \\ &\times \frac{\pi^3}{3024}\frac{k_BT}{\hbar}\left(\frac{\mu_B H}{E_t}\right)^2\left(\frac{k_BT}{E_t}\right)^6. \end{aligned} \tag{29}$$

The ratio of Raman and direct rates is of order $\Gamma_R/\Gamma_D \sim 10^{-2}(k_BT/\mu_BH)^2(k_BT/E_t)^4$. Consequently, Raman processes dominate over direct processes at high temperature and low field.

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